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Beryllium 10/beryllium 7 as a tracer of stratospheric transport

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Abstract. We use the ratio between cosmogenic radionuclides beryllium 10 ($t_{1/2}$ = 1.5 × 10^6 years) and beryllium 7 ($t_{1/2}$=53 days) to study stratospheric transport, particularly the exchange between the tropics and the high latitudes and as an indicator of stratosphere-troposphere exchange. Both tracers are generated steadily, with maximum production occurring at midlatitudes of the stratosphere. During transport away from their source the ratio $^{10}Be/^{7}Be$ increases and thus acts as a clock for air mass age. According to model (GISS GCM) calculations, the tropics is a reservoir of high-ratio air with the high magnitude of $^{10}Be/^{7}Be$ partly a consequence of air mixed in from higher latitudes. The observations of *Dibb et al.* [1994] show a vertical maximum in $^{10}Be/^{7}Be$ in the lower arctic stratosphere during the spring of 1989; model results qualitatively reproduce this maximum and suggest that it may result from transport of tropical air to high latitudes during winter. The model ratio values are not so large as in these observations, possibly due to either excessive leakage into the troposphere near the pole or to insufficient transport from the tropical stratosphere to the pole. Beryllium 10 and ^{7}Be observations, combined with model analysis, can be a useful indicator of transport processes in the troposphere/stratosphere system.

1. Introduction

The current picture of stratospheric dynamics includes influx from the troposphere and ascension of air in a relatively isolated "tropical pipe" region [e.g., *Plumb,* 1996], efficient meridional mixing by planetary waves in a midlatitude "surf zone," and isolation of poles during the winter in polar vortices. Various tracer studies have sought to quantify the degree of isolation of the tropics and of the polar vortices. Entrainment of extratropical air into the tropical pipe is a means of lofting aircraft exhaust into the upper stratosphere. The mixing rate of tropical air into the extratropics is also important for determining the sensitivity of midlatitude ozone loss to industrial pollutants. Furthermore, the amount of penetration of the polar vortices by tropical and midlatitude air has important implications for the study of ozone hole chemistry and morphology.

Recent studies of tropical chemistry indicate that there is 30-50% entrainment of extratropical air into the tropical pipe [*Boering et al.,* 1996; *Volk et al.,* 1996], with a minimum of mixing occurring around 19-21 km [*Minschwaner et al.,* 1996]. "Leakage" out of the tropical pipe is also apparently substantial and was evident, for example, from the outflow of N_2O and water [*Randel et al.,* 1993]. The outflow rate is faster, with a timescale of about 6 months as compared to a timescale of about 13.5 months for entrainment [*Volk et al.,* 1996] but may decrease with altitude [*Volk et al.,* 1996; *Boering et al.,* 1996].

Several studies have also revealed penetration of the polar vortex by lower-latitude air. For example, *Plumb et al.* [1994] demonstrated that intrusion into the lower arctic polar vortex accompanies tropospheric meteorological disturbances and found about 5% intrusion during one month. *Waugh et al.* [1994] simulated the transport of tropical air rich in N_2O and water as it penetrated the polar vortex during wave-breaking events. These studies emphasize the fine scale of these phenomena and point out the difficulty of coarse scale observations and models in capturing them.

Cosmogenic radionuclides, with their unique source and sink characteristics, provide a perspective on stratospheric dynamics which is complementary in many respects to the tracers currently used. Several previous studies have reported observations of cosmogenic radionuclides and explored their utility as stratospheric

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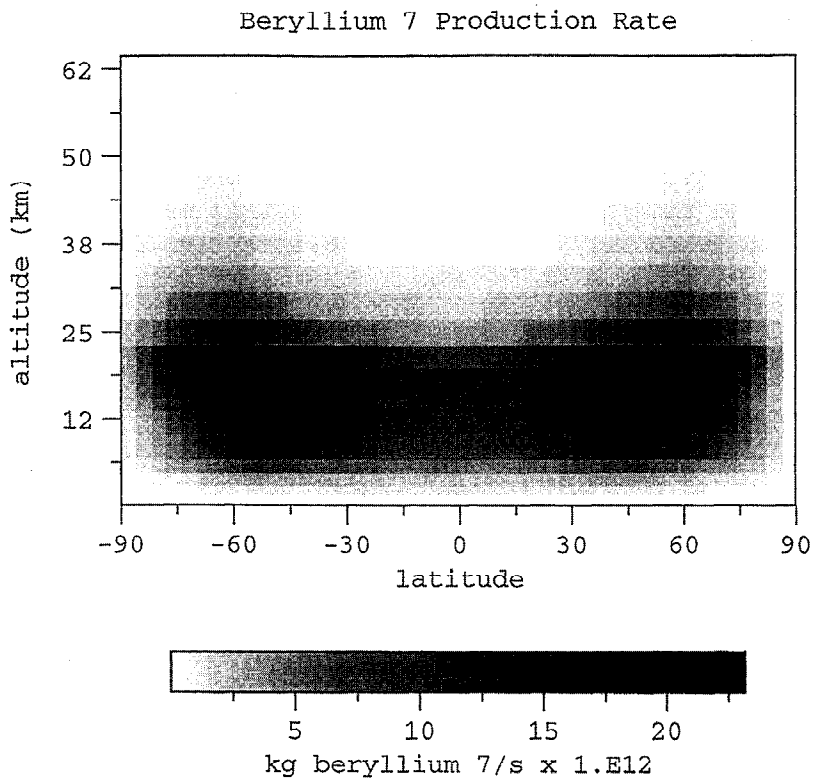

Figure 1. Zonal total, annual average ^{7}Be source [from *Lal and Peters*, 1967]. Production does not vary significantly with longitude. The ^{10}Be production is proportional to ^{7}Be.

tracers [e.g., *Bhandari and Rama*, 1963; *Bhandari et al.*, 1966]. These studies demonstrated that ratios of these tracers with different half-lives can be used to clock air mass age. Here we work with ^{7}Be and ^{10}Be which have identical source distributions (Figure 1). Production results from cosmic ray impact on N and O atoms and is highest at midlatitudes of the stratosphere. The production rate is nearly steady, although it varies by about 20% from the mean due to fluctuations in solar activity [e.g., *Koch et al.*, 1996; *Koch and Mann*, 1996; *Beer et al.*, 1990; *Lal and Peters*, 1967], and beryllium is chemically nonreactive. The production distribution makes the cosmogenic radionuclides suitable for studying meridional transport from midlatitudes to the poles or to the tropics and for studying stratosphere-troposphere exchange. Beryllium 7 is lost by radioactive decay, with a half-life of 53 days; decay of ^{10}Be is negligible relative to atmospheric residence times, with a half-life of 1.5×10^{6} years. During transport away from the regions of highest production, ^{7}Be decays and the ratio $^{10}Be/^{7}Be$ increases. The ratio $^{10}Be/^{7}Be$ therefore acts as a clock indicating the path-integrated age of the air mass. In addition to decay of ^{7}Be, both ^{7}Be and ^{10}Be are lost to precipitation scavenging in the troposphere, since they are associated with aerosols. Most of the tracer mass which enters the troposphere is rained out and does not reenter the stratosphere.

Beryllium 7 measurements are based on gamma decay and are relatively plentiful (e.g., referenced by *Koch et al.* [1996]). However, ^{10}Be measurements require accelerator mass spectrometry, are expensive, and therefore scarce. Among the only high-altitude ^{10}Be observations are reported by *Dibb et al.* [1994] and *Raisbeck et al.* [1981]. These observations indicate a vertical maximum in the ratio $^{10}Be/^{7}Be$, peaking above the arctic tropopause. These studies speculated that this high ratio might be a consequence of the slow subsidence of air from aloft.

We use the Goddard Institute for Space Studies general circulation model (GISS GCM) to further investigate the utility of the ratio $^{10}Be/^{7}Be$ as a stratospheric tracer. In particular, the model is used to determine the mechanism responsible for the high ratio above the arctic tropopause. Is the maximum a consequence of the isolation and aging of air above the pole, or does it result from the transport of high-ratio air originating at lower latitudes? Leakage of tracer into the troposphere also affects the distribution of $^{10}Be/^{7}Be$. Comparison of model results with high-altitude and surface observations allows us to check the model's ability to simulate stratospheric transport and stratosphere-troposphere exchange.

Beryllium 10 and ^{7}Be were modeled previously by *Rehfeld and Heimann* [1995] in a 19-layer version of the ECMWF model. They used the ratio $^{10}Be/^{7}Be$ pri-

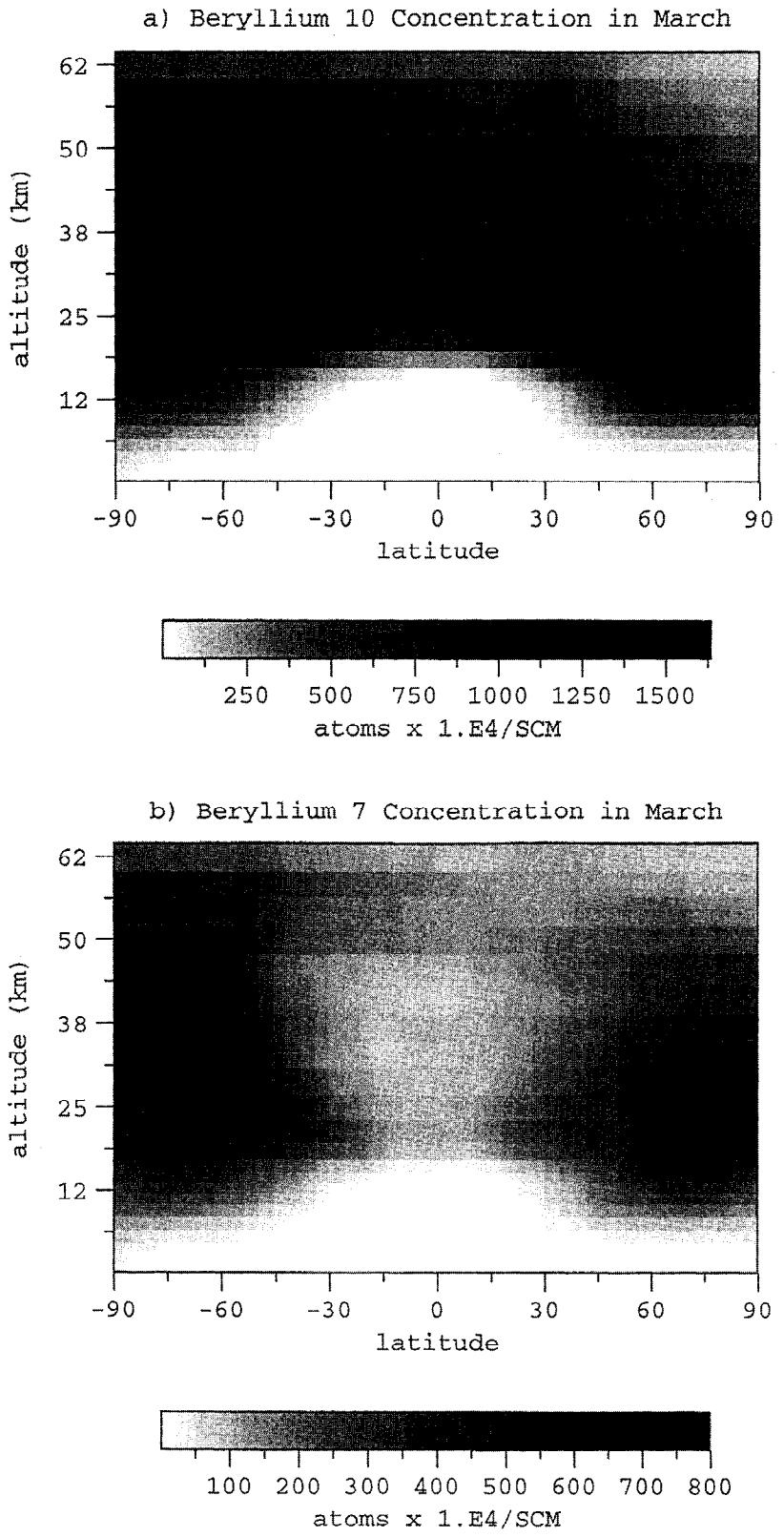

Figure 2. Zonal average (a) ^{7}Be and (b) ^{10}Be model concentrations for March in the lowest 25 layers of the 31-layer model.

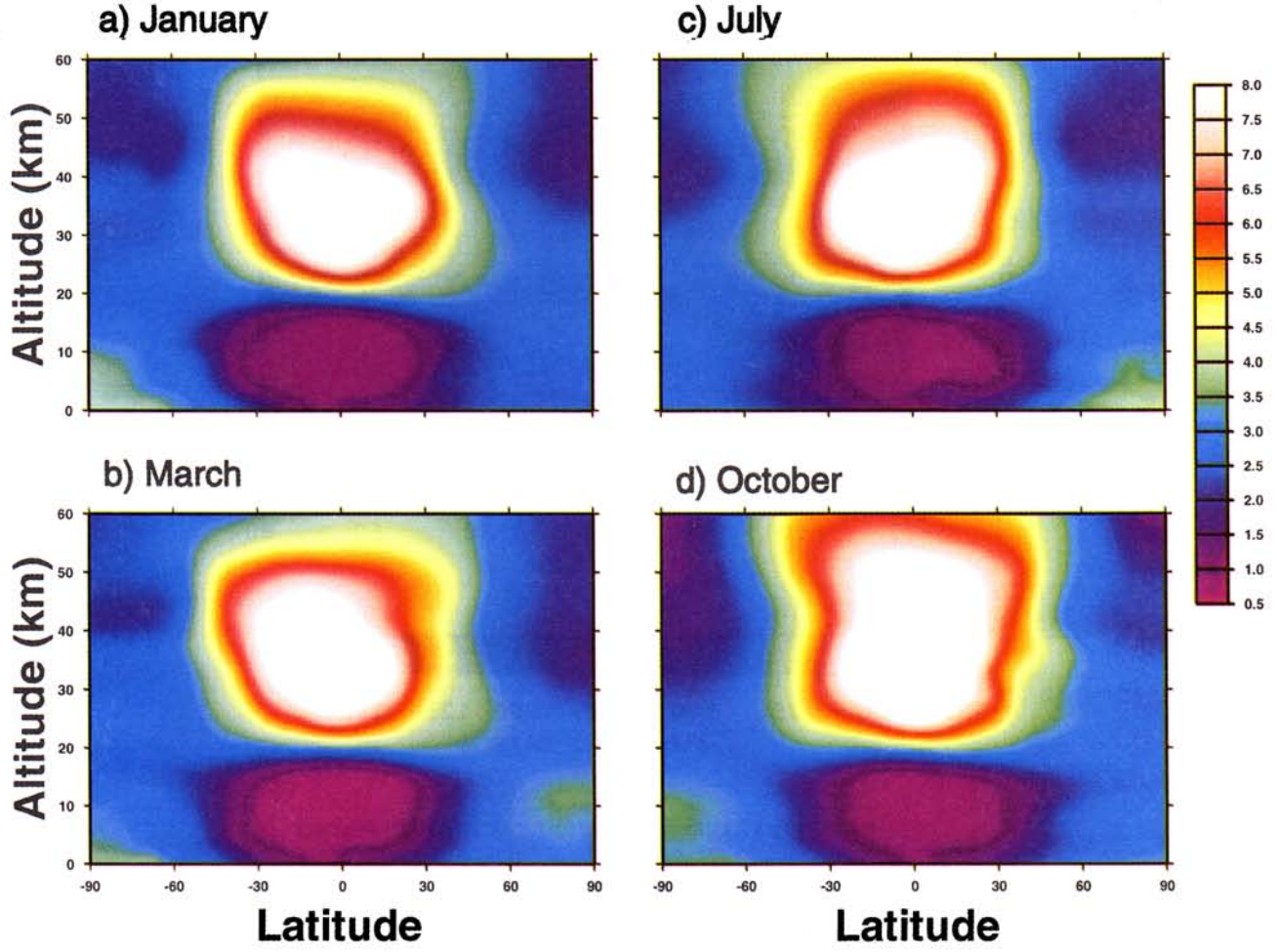

Plate 1. Zonal average ratio ^{10}Be/^{7}Be in (a) January, (b) March, (c) July, and (d) October.

marily as an indicator of stratosphere-troposphere exchange. We will compare the GISS results with their model results.

2. Model

We use 31-layer and 18-layer versions of the GISS GCM to simulate ^{7}Be and ^{10}Be (both have $\approx$ 12 layers in the troposphere). Horizontal resolution is 4° by 5°. The top of the 31-layer model is 0.003 mbar and the top of the 18-layer model is 10 mbar. All results presented are from the 31-layer model, except for the "limited source" experiments, described below. Since the top of the 18-layer model is low, we also use the 31-layer model to distinguish dynamical features from model-top effects. We allow 5 years of spin-up and show results from year 6. In comparison with the older version of the GISS GCM [*Hansen et al.*, 1983] this version has improved subroutines for the boundary layer, convection, land surface, a cloud liquid water budget, and uses the quadratic upstream scheme for heat and moisture advection as well as a fourth-order advection scheme. Details on the various components are discussed or referenced by *Rind and Lerner* [1996].

Figure 1 shows the ^{7}Be source [*Lal and Peters*, 1967]. The production rate of ^{10}Be relative to ^{7}Be is somewhat uncertain, with estimates ranging from 0.5 to 1.3 (see *Dibb et al.* [1994] for further discussion). As pointed out by *Dibb et al.* [1994], the production ratio must be lower than the minimum observed values, which are less than 0.7. We use a ^{10}Be production rate that is 0.55 that of ^{7}Be [*Lal and Peters*, 1967], and this is also within the range favored by *Dibb et al.* [1994] (0.6 $\pm$ 0.1). In addition to sinks from radioactive decay and deposition in the troposphere, aerosol settling can be an important sink for the long-lived ^{10}Be above the middle stratosphere. To include settling in the model, we increase the downward tracer mass flux using the settling velocity given by *Kasten* [1968]:

$$w = \frac{2}{9}(\rho_p r^2 g/\eta)[1 + \lambda/r(A + B\exp(-Cr/\lambda)] \quad (1)$$

where ρ_p is the particle density (assumed to be 1.8 g/cm^3, appropriate for sulfuric acid particles), r is the particle size, η is air viscosity, λ is the mean-free path of air, and A, B, and C are empirical constants found to be 1.249, 0.42, and 0.87, respectively. We assume that particles have a radius of 0.1 microns, typical of the mean radius of background aerosols observed in the middle to upper stratosphere [*Hofmann*, 1990]. Since

the mean-free path of air is large relative to aerosol particle size in the middle and upper stratosphere, settling rates (e.g., approximately 3, 30, and 300 km/year at 28, 45, and 62 km, respectively) can be significant. As discussed below, we neglect settling in the 18-layer experiments.

Figure 2 shows the zonal-average ^{7}Be and ^{10}Be model concentrations for March in the 31-layer model. Relatively slow transport occurs during the summer and fall, allowing large concentrations to build up near the pole (south pole in Figure 2). Tropospheric concentrations are low due to precipitation scavenging. This precipitation scavenging assures that very little tracer reenters the stratosphere.

Plate 1 shows the ratio ^{10}Be/^{7}Be for four different months. The tropical troposphere has very low ratio because convective precipitation removes both tracers, and the production "clock" is reset to zero, and because stratospheric input mostly occurs at higher latitudes. The ^{10}Be/^{7}Be ratio is highest in the tropical stratosphere where production is low (see Figure 1), and transport from high production regions allows decay of ^{7}Be (compare Figures 2a and 2b). Note the vertical maximum in ^{10}Be/^{7}Be during March in the arctic lower stratosphere. This ratio maximum above the arctic tropopause begins during the winter, strengthens during the spring, then fades during the summer and fall. In the arctic troposphere the ratio is maximum during the spring and summer. A similar feature appears in the model over the south pole during austral spring.

The ECMWF simulation of ^{10}Be and ^{7}Be [*Rehfeld and Heimann,* 1995] does not appear to have a vertical maximum in ^{10}Be/^{7}Be over the arctic (this may be due to lack of contour resolution in the figures). The ratio seasonality in the arctic peaks in winter near the tropopause (earlier than the GISS model) and during the spring at the surface (similar to the GISS model).

3. Model and Observations in the Arctic Stratosphere

Figures 3 and 4 compare zonal average vertical profiles from the model with high-altitude observations [*Dibb et al.,* 1994]. Figure 3 has midlatitude (30°N-56°N) observations from the PEM West A mission during fall 1991. Note that a large range of ^{10}Be/^{7}Be exists for these latitudes in the model (indicated by the bars across the model profile; see also Plate 1) and thus agreement with observations is good.

Figure 4 has higher-latitude (65°N to 82°N) observations, including one set from July 1978 [*Raisbeck et al.,* 1981] and another from March 1989 [*Dibb et al.,* 1994]. The former were derived from the Environmental Measurements Laboratory (EML) high-altitude balloon-sampling program, which had the purpose of monitoring atmospheric radioactivity. The latter samples were collected during the Arctic Gas and Aerosol Sampling Program (AGASP 3), which focused primarily on tropospheric arctic haze chemistry and transport. In all cases the ^{10}Be measurements were made on the same samples from which the ^{7}Be values were derived. Model profiles are shown for 74°N during March and July. The model ^{10}Be and ^{7}Be are somewhat higher than observed just above the tropopause. The July profiles agree reasonably well with the three higher-level observations from July. The model ratio ^{10}Be/^{7}Be in March does have a maximum above the tropopause; however, it is not so high as observed. The model maximum disappears by July rather than persisting into the summer as the one lower stratosphere measurement in July suggests it should. Note that these high-ratio values (greater than 4) observed in the lower stratosphere exist in the model only in the tropics.

In summary, the model does simulate a maximum in ^{10}Be/^{7}Be in the lower arctic stratosphere during the winter and spring, although it is smaller than observed. Next, we present model experiments designed to investigate the source region responsible for the arctic maximum.

4. Limited Source Experiments

To investigate the origon of the ^{10}Be/^{7}Be maximum in the arctic, we conducted 18-layer model experiments in which we zeroed the radionuclide source everywhere except in select regions. So, in addition to running the model with the full source, we also did one experiment with the source set to zero everywhere except in the tropics (between 24°N and 24°S), and another with the source zero everywhere except near the poles (poleward of 60°). We call the tropical and polar source experiments TS and PS, respectively. To find the contribution from the midlatitude source (MS) we take the difference as follows: MS = full source run - TS - PS. Note that the source is not evenly distributed among the three regions. The tropical, midlatitude and polar sources comprise 22%, 53%, and 25%, respectively, of the total source.

4.1. Origin of High Ratio in the Arctic

In Figure 5 we show model profiles in the arctic during March from each of the three limited source experiments and the full-source run. Note that much of the ^{10}Be at the pole is derived from midlatitudes, although all three source regions contribute significantly to its total. Beryllium 7, which decays rapidly, has much of its source locally, near the pole. Figure 5c shows that the ^{10}Be/^{7}Be ratio from the tropical source is quite large. The (full source) model ratio in the arctic results from some mixture of the polar source and the sources at lower latitudes. However, in order to get a ratio as high as was observed above the tropopause, a greater contribution from the tropical source is required.

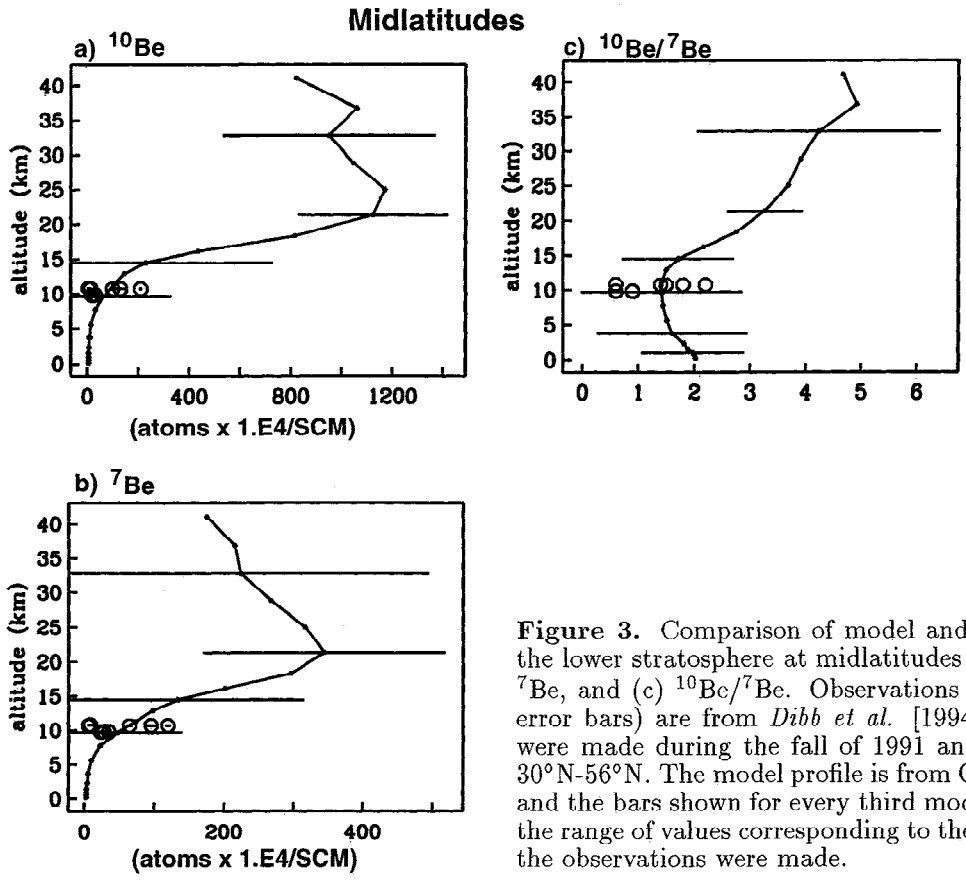

Figure 3. Comparison of model and observations in the lower stratosphere at midlatitudes for (a) ^{10}Be, (b) ^{7}Be, and (c) $^{10}Be/^{7}Be$. Observations (hexagons, with error bars) are from *Dibb et al.* [1994]. Observations were made during the fall of 1991 and span latitudes 30°N-56°N. The model profile is from October at 40°N, and the bars shown for every third model layer indicate the range of values corresponding to the latitudes where the observations were made.

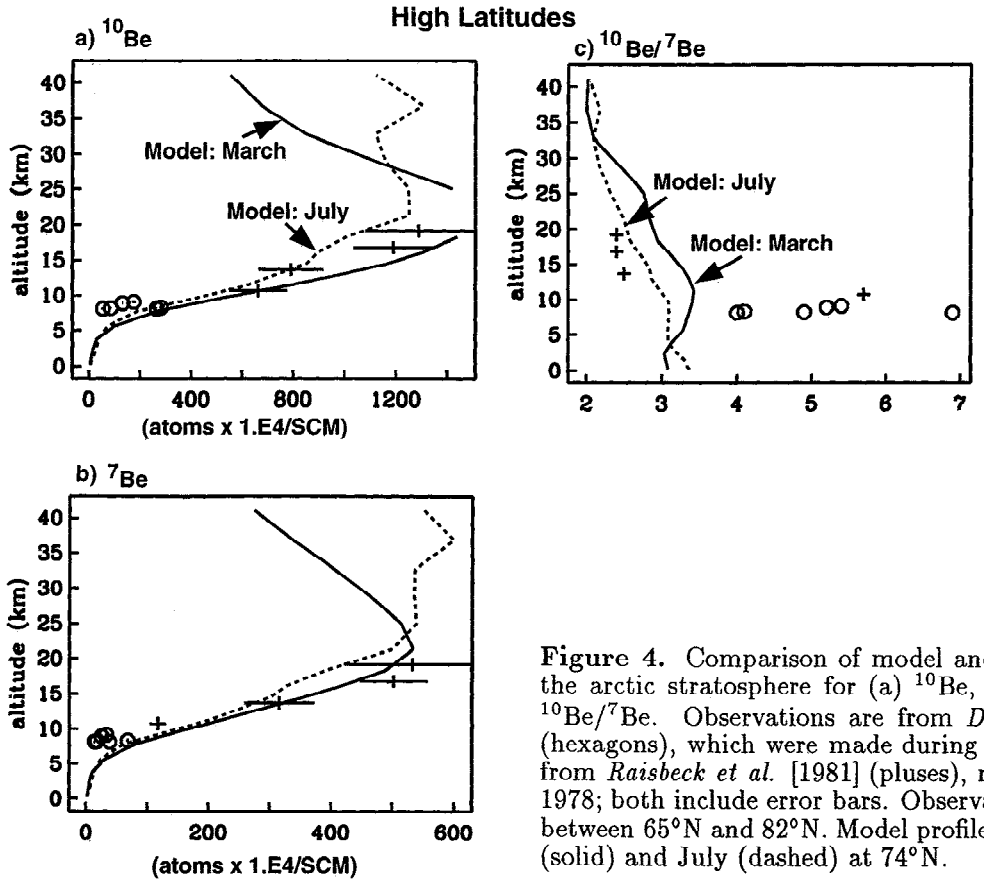

Figure 4. Comparison of model and observations in the arctic stratosphere for (a) ^{10}Be, (b) ^{7}Be, and (c) $^{10}Be/^{7}Be$. Observations are from *Dibb et al.* [1994] (hexagons), which were made during March 1989, and from *Raisbeck et al.* [1981] (pluses), made during July 1978; both include error bars. Observations were taken between 65°N and 82°N. Model profiles are from March (solid) and July (dashed) at 74°N.

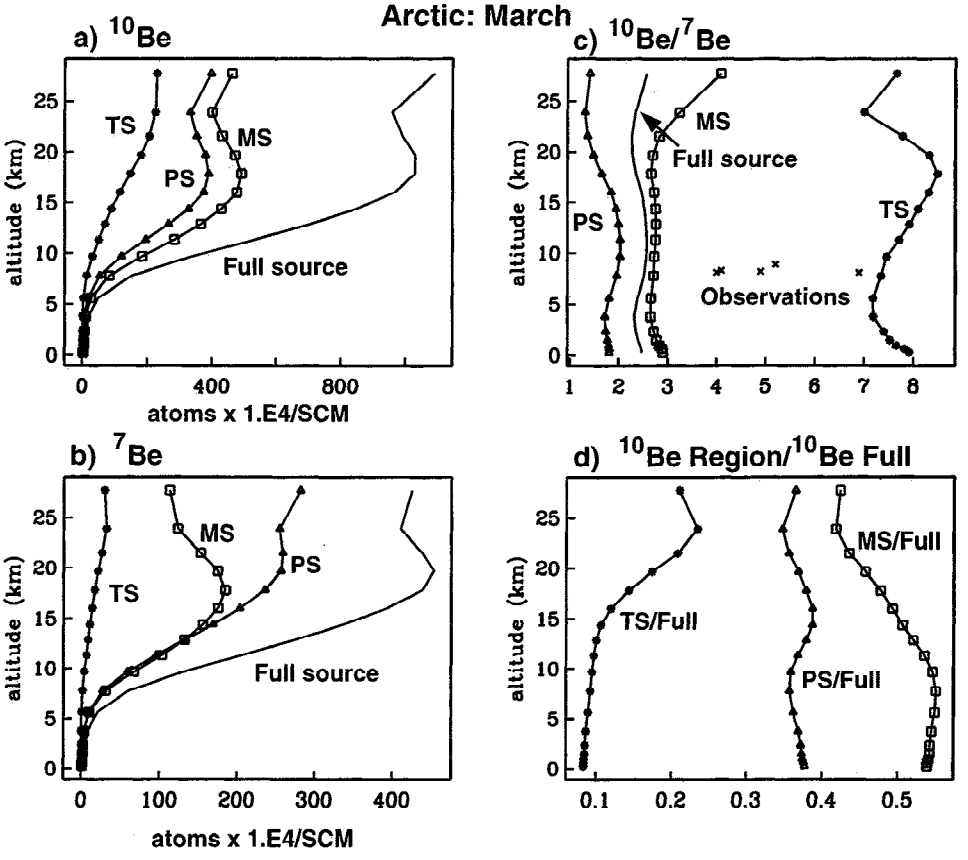

Figure 5. Limited source model profiles at 74°N in March, showing results from the full source (solid curve), the midlatitude source (MS) (squares), the polar source (PS) (triangles), and the tropical source (TS) (stars) for (a) ^{7}Be, (b) ^{10}Be, and (c) $^{10}Be/^{7}Be$. (d) ^{10}Be from each source region normalized by the full source. Observed ratio values are also shown in Figure 5c.

As mentioned earlier, we have not included aerosol settling in the 18-layer model. This assumption can be tested by comparing Figures 5a and 4a. In a previous 31-layer model run which did not include settling, we found that ^{10}Be concentrations became much larger than observed, particularly at the highest altitudes. Aerosol settling reduced the ^{10}Be concentrations to be consistent with observations (Figure 4a). In the 18-layer model (Figure 5a), even without settling, ^{10}Be does not quite reach the observed levels; thus settling is not required to prevent buildup of ^{10}Be. This may be due to the truncation of the source by the lower model top (around 30 km; see Figure 1).

We can get a rough estimate of the amount of transport from each region to the arctic by considering the amount of ^{10}Be transported, since ^{10}Be decay is small. Figure 5d shows the ^{10}Be in each source region divided by the full-source ^{10}Be. From this we see that $\approx$10% of the model ^{10}Be above the polar tropopause has its source in the tropics. Note that the amount of ^{10}Be is not the same as the amount of air since the ^{10}Be source is not evenly distributed in the stratosphere. However, since the radionuclide production is relatively small in the tropics, the percent of ^{10}Be derived from the tropics will be lower than the percent of air.

4.2. Origin of High Ratio in the Tropics

Next, we use the limited source experiments to determine the origin of the high ratio in the tropics. Is the high ratio a consequence of the isolation of radionuclides generated within the tropical pipe, or is it a result of the transport of air (with large decay of ^{7}Be) from higher latitudes? Figure 6 shows that much of the ^{10}Be and ^{7}Be in the tropics has its source in the tropics. Figure 6c shows that the ratio $^{10}Be/^{7}Be$ from the tropical source is relatively high in the stratosphere but not so large as what appears in the full run. In order to get the full-run ratio magnitude, some air, which has passed through the higher latitude sources, is required. Thus we have evidence of transport into the model's tropical pipe. Again, we can estimate the amount of transport into the tropical stratosphere in the model by considering the ^{10}Be contribution from each source region (Figure 6d). Approximately 50% of the ^{10}Be in the tropics comes from higher latitudes. Actual air transported into the tropics would be lower since the source distribution is weighted toward the higher latitudes.

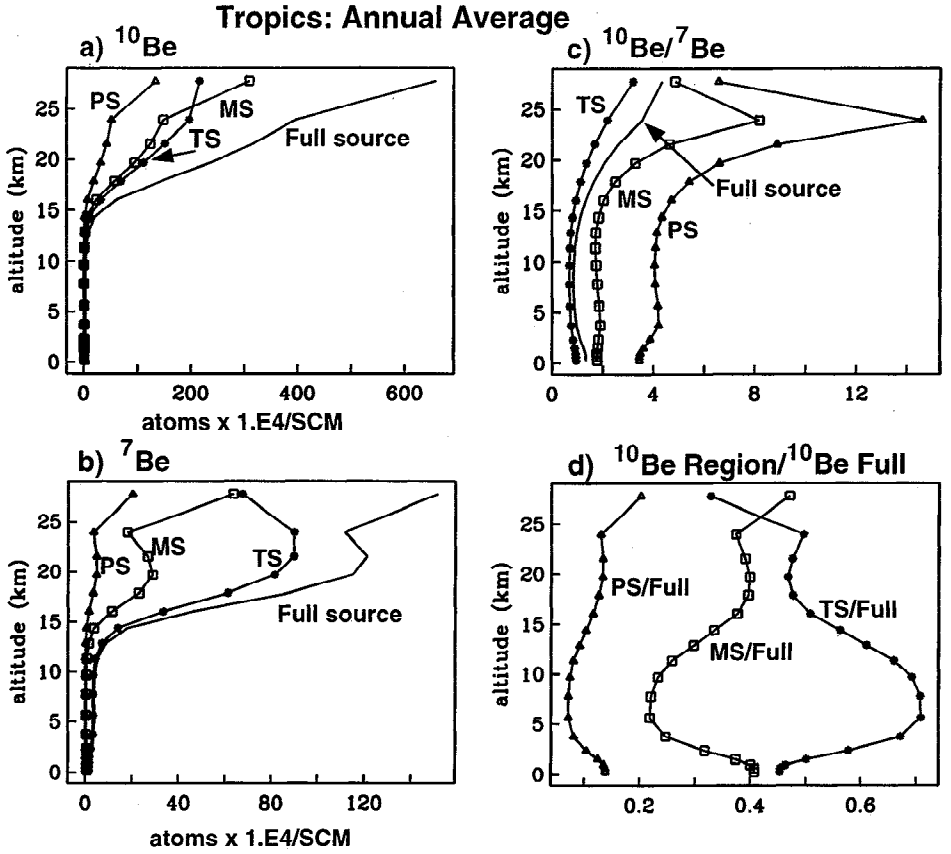

Figure 6. Same as Figure 5 for annual average profiles in the tropics.

5. Poleward Transport of High-^{10}Be/^{7}Be Air

The model has a seasonal pattern in which high-ratio air appears over the arctic during late winter, increases in magnitude during the spring, and fades by summer. Large poleward transport and a maximum in the ^{10}Be/^{7}Be ratio in the lower arctic stratosphere occurs each winter in the model. Figure 7a shows zonal average ^{10}Be transport during January. Note the large poleward transport from the tropics to the pole in the lower stratosphere. This transport is enhanced by mid-latitude subsidence from aloft.

Vigorous transport in the northern hemisphere is limited to the winter and spring. As shown in Figure 7b, transport during the (northern hemisphere) summer is slow and equatorward. Figure 7b also shows that transport during the southern hemisphere winter is less vigorous than in the northern hemisphere winter (Figure 7a).

As we have shown, the model ^{10}Be/^{7}Be ratio maximum in the northern hemisphere springtime is not so high as observed, and it fades during the summertime. It is possible that poleward transport in the model is not large enough. Another possibility is that excessive leakage into the troposphere does not permit the ratio to build up to the observed values.

6. Tropospheric ^{10}Be/^{7}Be As Indicator of Stratosphere-Troposphere Exchange

One way to test whether excessive leakage into the troposphere is depleting the ratio in the stratosphere is to compare the model ^{10}Be/^{7}Be ratio in the troposphere with surface observations. High ratios in the troposphere generally indicate a large stratospheric component. However, precipitation scavenging resets the air concentration ratio to its production value (0.55); therefore it is best to look for the stratospheric component either within rainwater (the deposition flux) or in air that has undergone little scavenging. In Table 1 we compare the deposition fluxes observed at three locations [*Monaghan et al.*, 1986; *Baskaran et al.*, 1993; *Knies*, 1994] with corresponding model values. (Beryllium 7 and ^{10}Be measurements are not necessarily from the same studies or years.)

In New Haven, Connecticut and College Station, Texas, the model deposition flux is lower than observed, for both ^{7}Be and ^{10}Be. Since both sites are coastal, the discrepancy may result from excessive amounts of oceanic air in the model (^{7}Be concentrations tend to be higher over land [e.g., *Koch et al.*, 1996]). The ratio ^{10}Be/^{7}Be agreement is reasonably good, though somewhat high at all three of the sites.

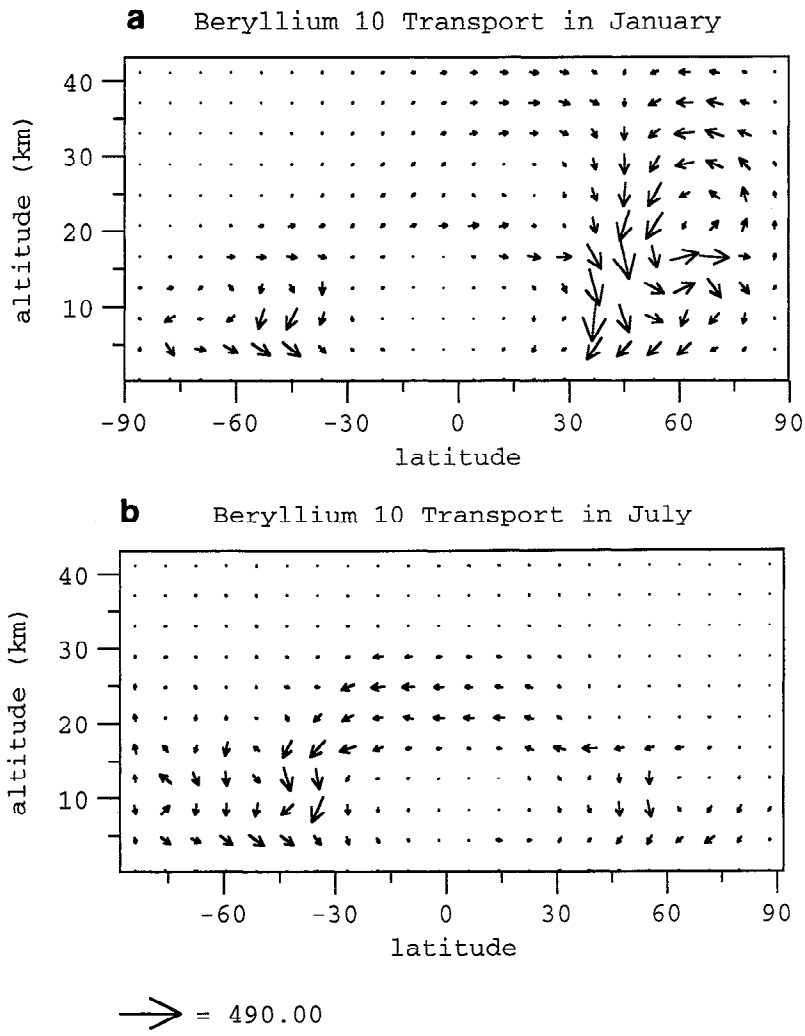

Figure 7. Zonal average ^{10}Be transport in (a) January and (b) July in kg $^{10}Be/s \times 10^{12}$.

Of greater interest to our current study is the ratio in the arctic troposphere. *Dibb et al.* [1994] observed ^{7}Be and ^{10}Be surface concentrations in Alert, Northwest Territories. We show these observations with the final three years of model results in Figure 8. Since the observations are from one year, we wrap them around to compare with successive model years. The ^{7}Be and ^{10}Be surface concentrations are observed to peak in the springtime, while the $^{10}Be/^{7}Be$ ratio peaks during the summer. The model seasonality has the opposite trend: the ^{7}Be and ^{10}Be concentrations peak during the summer, but the $^{10}Be/^{7}Be$ ratio peaks in the spring and summer. The model concentrations and ratio are greater than observed. The early peaking of the ratio in the model troposphere suggests that leakage into the troposphere also begins too early. (The slight upward drift in the model ratio is because the model ^{10}Be is approaching equilibrium.)

From these limited observations it does appear that leakage into the troposphere is somewhat excessive in the model, particularly at high latitudes. We might thus expect that the $^{10}Be/^{7}Be$ model ratio in the lower stratosphere would attain higher values if leakage into the troposphere were slower and later in the year, as the observations suggest it should be. A major caveat to these conclusions is that the observations are from one year only, and it is not known how representative the results are of climatological conditions.

Table 1. Model Versus Observed Deposition Flux

Site	Species	Observed	Model
New Haven, Connecticut	^{10}Be	307.0	277.0
	^{7}Be	141.0	119.0
	$^{10}Be/^{7}Be$	2.2	2.3
College Station, Texas	^{10}Be	239.0	181.0
	^{7}Be	142.0	95.0
	$^{10}Be/^{7}Be$	1.7	1.9
Indiana	^{10}Be	218.0	317.0
	^{7}Be	119.0	140.0
	$^{10}Be/^{7}Be$	1.8	2.3

Atoms 1 x $10^{4}/cm^{2}/yr$.

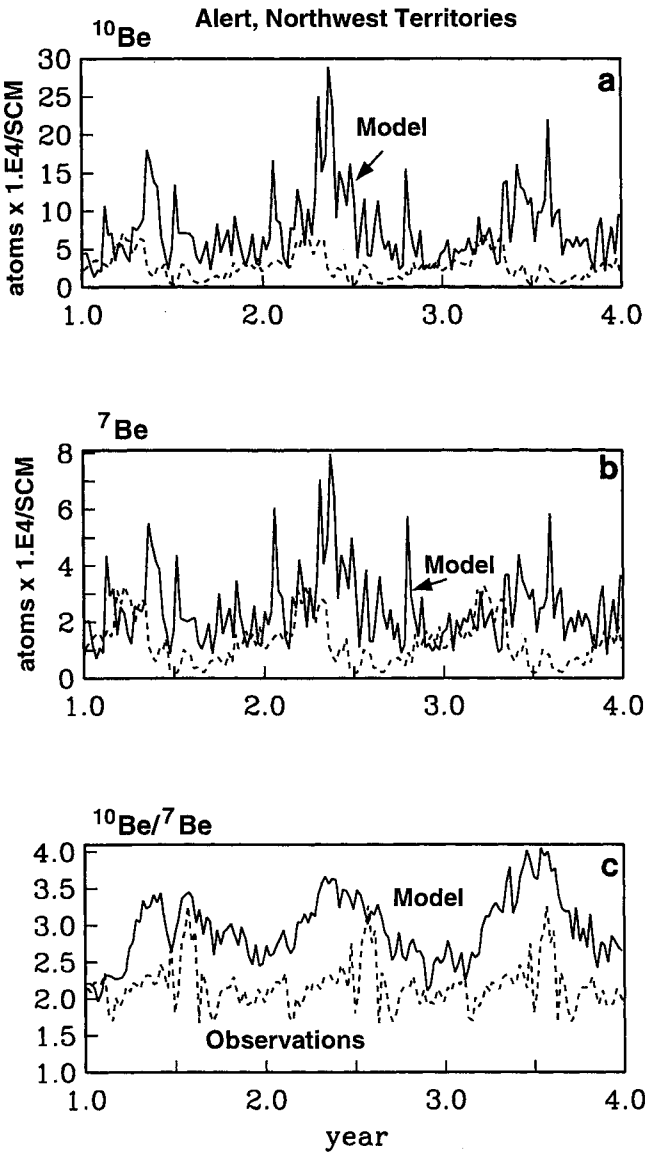

Figure 8. Observed (dashed) and modeled (solid) (a) ^{10}Be, (b) ^{7}Be, and $^{10}Be/^{7}Be$ from Alert, Northwest Territories. Observations from one year [*Dibb et al.*, 1994] are repeated for comparison with three model years.

7. Discussion and Conclusions

We have used a three-dimensional model to simulate and study the evolution of a maximum in the $^{10}Be/^{7}Be$ ratio in the lower arctic stratosphere. The model successfully generates the maximum during the winter and spring; however, the ratio is not so high as observed. We showed that increased transport of high ratio tropical air could increase the arctic ratio to the observed levels. The model already transports a significant amount from the tropics: 10% of the ^{10}Be at the pole had its source in the tropics. If we assume rapid transport and use a tropical ratio value of 10 and a background polar ratio value of 3.5, then we would need 40% of the polar air to be derived from the tropics in order to get the ratio as high as observed. This amount is much greater than the 5% transport from the tropics in one winter month found by *Plumb et al.* [1994].

The model indicates that the $^{10}Be/^{7}Be$ ratio is quite high in the tropical stratosphere. According to the model, about 50% of ^{10}Be in the tropics is entrained from higher latitudes, an amount at the upper range of previous estimates [e.g., *Boering et al.*, 1996; *Volk et al.*, 1996]. There are no measurements of ^{10}Be in the tropical stratosphere, and these are needed to allow us to use the model to estimate transport into and out of the tropics. It is also possible that a higher model ratio in the tropics is required to generate sufficiently high values in the arctic lower stratosphere.

The high-ratio observations in the lower arctic stratosphere were made during the spring of 1989 [*Dibb et al.*, 1994], the year of a major polar warming event. The polar warming might have caused anomalously high poleward transport and hence high ratios. However, one additional sample having a high ratio was collected in July 1978 [*Raisbeck et al.*, 1981], after a winter without a major warming. Further observations are required to learn whether this ratio appears every spring and how long it persists. The model maximum begins during late winter, strengthens during the spring, and then fades during the summer. Compared with one year of surface observations in the arctic, it appears that model stratosphere-troposphere exchange is high and begins too early. However, compared with $^{10}Be/^{7}Be$ observations in rainwater at three (lower latitude) sites, the model stratosphere-troposphere exchange is reasonably well simulated. Nevertheless, it is possible that excessive springtime leakage into the troposphere may deplete the ratio in the lower stratosphere so that it does not build to the values observed or persist into the summer. Further tropospheric measurements of $^{10}Be/^{7}Be$ are required to rigorously test model stratosphere-troposphere exchange.

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